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RADIOGRAPHY WITH THE FISSION NEUTRONS FROM CALIFORNIUM-252

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MATERIALS SCIENCES DIVISION

July 1977

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ARMY MATERIALS AND MECHANICS RESEARCH CENTER
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REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM	
1. REPORT NUMBER AMMRC TR 77-18		2. GOVT ACCESSION NO.	
3. TITLE (and Subtitle) RADIOGRAPHY WITH THE FISSION NEUTRONS FROM CALIFORNIUM-252		3. RECIPIENT'S CATALOG NUMBER	
4. AUTHOR(s) John J. Antal and Robert L. Becker*		5. TYPE OF REPORT & PERIOD COVERED Final Report	
6. PERFORMING ORGANIZATION NAME AND ADDRESS Army Materials and Mechanics Research Center Watertown, Massachusetts 02172 DMXMR-D		6. PERFORMING ORG. REPORT NUMBER	
7. CONTROLLING OFFICE NAME AND ADDRESS U. S. Army Materiel Development and Readiness Command, Alexandria, Virginia 22333		8. CONTRACT OR GRANT NUMBER(s)	
8. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS D/A Project: 1T161102AH42 AMCMS Code: 611102.11.85500 Agency Accession: DA OC4683	
9. DISTRIBUTION STATEMENT (of this Report) Approved for public release; distribution unlimited.		12. REPORT DATE July 1977	
10. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)		13. NUMBER OF PAGES 10	
11. SUPPLEMENTARY NOTES Consultant, Professor, Boston College		15. SECURITY CLASS. (of this report) Unclassified	
12. KEY WORDS (Continue on reverse side if necessary and identify by block number) Radiography Fast neutrons Imaging techniques Track-etching Cellulose nitrate Californium-252		15a. DECLASSIFICATION/DOWNGRADING SCHEDULE	
13. ABSTRACT (Continue on reverse side if necessary and identify by block number)			
(SEE REVERSE SIDE)			

Block No. 20

ABSTRACT

Fission neutron radiography, with images formed on thin sheets of cellulose nitrate, has been investigated using neutrons from Cf-252. A polyethylene converter provides recoil protons from neutron elastic scattering by hydrogen, which in turn create damage in the cellulose nitrate. Chemical etching then produces a frosted etch-track image which can be reproduced photographically by either scattered or transmitted light. The most readily available cellulose nitrate sheets were found to contain undesirable internal defects and thus films are recast in thicknesses of several mils. The images are of high resolution, as expected from a computer calculation which considered the direction and ranges of the recoil protons in polyethylene. All materials, including those of low atomic weight, may be radiographed with good penetration. The technique is simple and employs inexpensive materials.

INTRODUCTION

Techniques are well established for performing neutron radiography using thermal neutrons from a nuclear reactor. The availability of ^{252}Cf , which provides an intense but portable source of neutrons, has stimulated interest in the application of these sources to neutron radiography in situations where a reactor is not available.

Neutron radiography using ^{252}Cf is usually accomplished with the source inside a moderator. Radiographs are taken with a beam of thermal neutrons that is extracted through a collimator, and the same techniques that are used at a reactor can be applied to reproduce the neutron image.

It would also be desirable that a radiography technique be developed which employs the fission neutrons directly from the ^{252}Cf source.¹ Some obvious advantages of fission neutron radiography are that the loss of neutrons during the moderation process is avoided, and perhaps more important, the essential point source of neutrons is maintained, so that collimation of the neutron direction is not needed. The resulting source facility is greatly simplified without a moderator or collimator.

It is necessary to realize that fast neutrons interact with matter differently than do thermal neutrons, and thus we are considering a somewhat different kind of radiography.²⁻⁴ Fast neutron interactions vary gradually and systematically with the mass number of the scattering nucleus, while thermal neutron cross sections can change dramatically from one element to the next. Fast neutrons can penetrate well through all materials, and could be used to examine details of any object, no matter what it is composed of or contained in. While fast neutron radiography is not as sensitive to such elements as hydrogen or boron, it can be used for detecting them and for examining the interior of structures containing the elements.

A major difficulty in fission neutron radiography has been in finding a suitable detector for the fast neutrons. This detector should be insensitive to the thermal neutrons and gamma rays which invariably are present along with the fast neutrons. The etch-track technique^{5,6} using cellulose nitrate has been successfully employed here for the detection of recoil protons from elastic neutron scattering in hydrogen. We have found this to be insensitive to the other radiations, particularly gamma rays, and to provide radiographs of exceptionally good resolution.

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6. FLEISCHER, R. L., PRICE, P. B., and HUBBARD, E. L. *Criterion for Registration in Dielectric Track Detectors*. Phys. Rev., v. 156, 1967, p. 353.

CELLULOSE NITRATE DETECTOR

Nearly all detectors of fast neutrons make use of neutron elastic scattering by hydrogen, and observe ionization, scintillations, or some other effect produced by the recoil protons. In fact, it was the observation of recoil protons that led to the initial discovery of the neutron.

The etch-track technique, by which charged particles are detected through the radiation damage they produce in dielectric materials, is used most frequently to detect heavy ions.^{7,8} It has been used in neutron dosimetry^{9,10} and for neutron radiography, by using converter plates of fissionable materials.^{3,11,12}

A most sensitive dielectric material, and the only one which we have found to respond to ions as light as hydrogen, is cellulose nitrate.^{13,14} We have employed as a detector a sheet of polyethylene and a cellulose nitrate film held between two thin rigid plates. The polyethylene has a high density of hydrogen and serves as a converter plate to provide the recoil hydrogen ions.

The detection can be understood in the following way. The recoil hydrogen ions pass from the polyethylene into the cellulose nitrate film. As they travel through the film they produce radiation damage along their path. When the film is subsequently immersed in a caustic solution, pits will be etched into the surface at locations where the ions penetrated the surface either in entering or leaving the film. Such pits provide a record of the detection of the neutron.

In our application the neutrons are incident normal to the surface of the polyethylene. In center-of-mass coordinates the scattering of neutrons by hydrogen is nearly isotropic, but in the laboratory coordinates the recoil protons are directed predominately toward the cellulose nitrate film.

Other mechanisms by which ions can penetrate the surface of the cellulose nitrate have been considered. Of these, the exoergic $^{14}\text{N} (n,p) ^{14}\text{C}$ reaction in the cellulose nitrate is the most likely to occur. In our application it will produce significantly fewer pits than does elastic scattering from hydrogen because the reaction has a lower cross section, because nitrogen is present in much smaller amounts than hydrogen, and because the flux of thermal neutrons is much smaller than the flux of fast neutrons in the absence of a moderator.

7. FLEISCHER, R. L., PRICE, P. B., and WALKER, R. M. *Tracks of Charged Particles in Solids*. Science, v. 149, 1965, p. 383.
8. COMSTOCK, G. M., FLEISCHER, R. L., GIRARD, W. R., HART, H. R., Jr., NICHOLS, G. E., and PRICE, P. B. *Cosmic Ray Tracks in Plastics: The Apollo Helmet Dosimetry Experiment*. Science, v. 172, 1971, p. 154.
9. BECKER, K. *Nuclear Track Registration in Dosimeter Glasses for Neutron Dosimetry In Mixed Radiation Fields*. Health Physics, v. 12, 1966, p. 769.
10. KEHRER, M. L., and ROBINSON, J. E. *A Simple Method for Making Neutron Measurements*. Int. J. Appl. Radiation and Isotopes, v. 23, 1972, p. 141.
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13. BECKER, R. L., and ANTAL, J. J. *An Etch Track Detector for Neutrons*. Bull. Am. Phys. Soc., v. 16, 1971, p. 1418.
14. CARPENTER, B. S., and LAFLEUR, P. D. *Observing Proton Tracks in Cellulose Nitrate*. Int. J. Appl. Radiation and Isotopes, v. 23, 1972, p. 157.

Recoil ^{12}C ions will be observed infrequently because of their short range. All possible (n, α) reactions and the endoergic (n,p) reactions will be observed infrequently because of their small cross section and small range of the emitted charged particles.

Our calculation of the probability that a neutron incident on the polyethylene will produce a recoil proton with range and direction so that it can penetrate into the cellulose nitrate is plotted in the top curve of Figure 1, as a function of neutron energy. This curve gives an upper limit for the efficiency with which neutrons can be detected by this front surface of the cellulose nitrate film. Of course, neutrons can be detected as well at the back surface by recoil protons as they leave the cellulose nitrate film. Some of these protons will have traveled through the film, and others will have been generated by elastic scattering processes inside the cellulose nitrate itself.

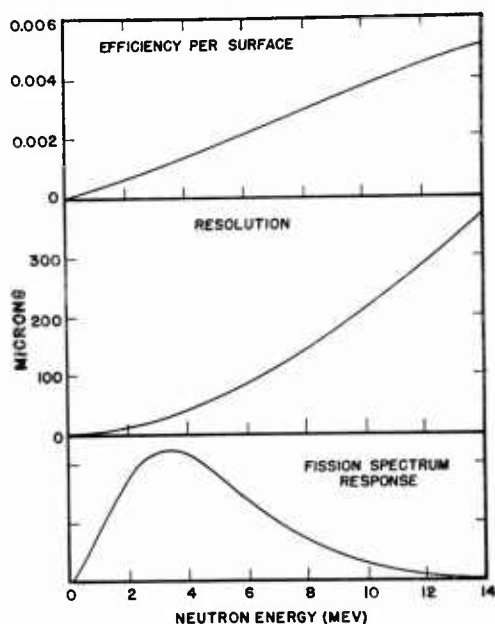


Figure 1. Calculated efficiency and resolution of cellulose nitrate as a proton detector (upper two curves). The bottom curve is obtained by folding the efficiency curve with a fission spectrum of neutrons to obtain the expected response of cellulose nitrate as a detector of fission spectrum neutrons.

19-066-1209/AMC-72

This calculated efficiency is seen to be a fraction of one percent, but increases with increasing neutron energy. The actual efficiency is observed to be smaller than the calculated value, implying that all recoil protons passing through the surface do not give observable pits.

The efficiency of detection depends on the component of the proton's displacement in a direction perpendicular to the film. The component parallel to the surface of the film will affect the resolution. The middle curve of Figure 1 shows the root mean square value of the transverse displacement as the recoil proton leaves the polyethylene. The averaging was taken over all possible scattering locations and all possible directions. This curve is a measure of the resolution inherent in cellulose nitrate as an imaging material. For the range of neutron energies which are available from ^{252}Cf , the intrinsic resolution is seen to be of the order of tens of microns.

Finally, the bottom curve of Figure 1 shows the relative response of this detector to the various neutron energies when a fission spectrum of neutrons is incident. To obtain this curve, the intensity of the fission spectrum at each energy is multiplied by the corresponding efficiency for detection as given in the top curve. This graph shows that more neutrons in the energy range between 3 and 4 MeV will be detected than in any other energy interval of equal width.

EXPERIMENTAL TECHNIQUES

Cellulose nitrate is not a precisely defined product. It exists in various degrees of nitration and polymerization, it may contain solvents or plasticizers, and it can exist in several physical forms. It is not yet known how to prepare the most sensitive material for etch-track use, but we have used materials from different sources with reasonably satisfactory results.

Uniform clear sheets or films are required for our purpose. Unfortunately, the commercially available sheets which we have tried have internal defects which are not apparent until the etching process removes the polish from the surface. The underlying striations in the film frequently obscure the image we are trying to observe. As a result we have cast our own films, using techniques similar to those described by Benton.¹⁵

We have successfully used two cellulose nitrate products.* Both give good quality images, although the sensitivity to radiation damage is not identical. The procedure is to dissolve the material in amyl acetate, then spread the solution on a glass sheet. The solvent is evaporated slowly, then removed as completely as possible by baking.

The neutron exposure produces no observable effect until the film has been etched. After the etching, which has usually taken place for 10 minutes in a 6.5 N solution of NaOH at 55 C, the etch pits resulting from radiation damage appear on the surface. These appear round when viewed under a microscope, with dimensions of 2 or 3 microns. When a sufficient density of pits is present, after an exposure of the order of 10^{11} neutrons/cm², the surface becomes fogged and an image is visible to the eye.

Prints from highly exposed films can be made in a photographic enlarger equipped with a point light source so that all rays of light are nearly axial. Regions corresponding to high pit density will receive less illumination and appear lighter in the resulting print.

When the exposure is significantly less than 10^{11} neutrons/cm², prints made in this fashion will have reduced contrast. In this case, good contrast may be achieved through use of dark field imaging, in which the print is exposed to light scattered by the film rather than transmitted through it.

15. BENTON, E. V. *A Study of Charged Particle Tracks on Cellulose Nitrate*. U. S. Naval Radiological Defense Laboratory Technical Report, USNRDL-TR-68-14, January 1968.

*Parlodion manufactured by Malinkrodt Chemical Co., and Type RS 5-6 SEC manufactured by Hercules, Inc.

Figure 2 illustrates the difference in appearance between dark field and the more conventional bright field imaging. This is one of our earliest exposures, using neutrons extracted through a 10-mm-diameter hole in the shipping container in which the source was transported. The image is of three cylinders: aluminum, plastic, and brass, which were placed near the axis of the hole. The top picture is the conventional view, in which the denser irradiation gives a brighter region on the print. The situation is reversed in the bottom picture.

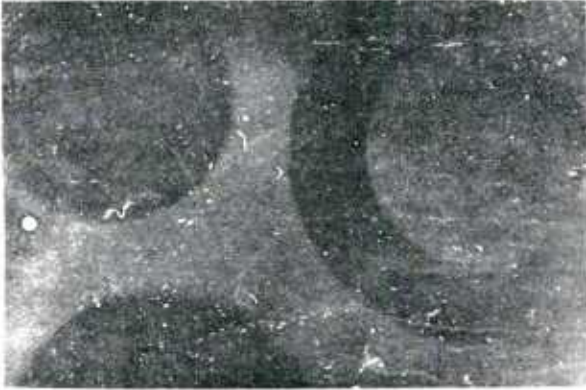


Figure 2. Bright (upper) and dark (lower) field reproductions of etch-track fission neutron radiographs. Most of the beam intensity is at the center of each picture. The three objects are cylinders of various materials.

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EXAMPLES OF RADIOGRAPHS

The radiographs which we present here were typically taken with a neutron exposure on the order of 2×10^{11} n/cm². For example, the film might have been about 20 cm from the 5-mg ²⁵²Cf source for a period of 30 hours. Some exposures were taken over the weekend, with a somewhat larger separation between source and film.

Figure 3 is a radiograph of a 6-ft steel measuring tape in a steel case. Individual turns of the tape and the steel return spring are resolved. These are of 0.13-mm thickness.

Figure 4 contains some electronic components. The glass envelope of the 6197 electron tube, as well as much of the internal structure, is well resolved. The pins of the tube, 1.0-mm-diameter, can also be seen. The remainder of the picture is a portion of a printed circuit. The circuit elements include transistors, capacitors, and carbon resistors. Along the right side of the picture is a delay line.

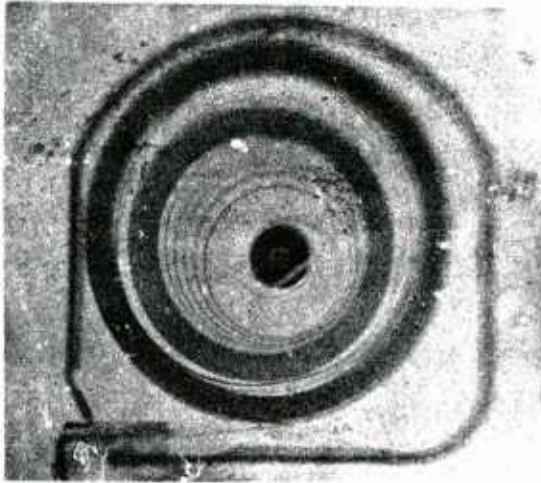


Figure 3. Fission neutron radiograph of a six-foot steel measuring tape wound in its steel case; the 0.13-mm (0.005") tape thickness is readily resolved.

19-066-1212/AMC-72

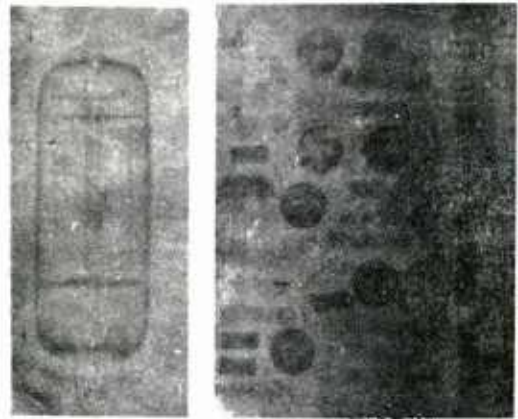


Figure 4. Fission neutron radiograph of a subminiature vacuum tube (left) and a printed circuit board (right) containing resistors, capacitors, transistors, and a delay line.

19-066-1208/AMC-72

Figure 5 contains two types of potentiometers. The outside cases are of cadmium-plated steel. The carbon element and phenolic support of the smaller potentiometer are easily seen, and the wiper arm can be observed on the original print. The larger potentiometer is wire-wound, and the phenolic ring on which the wires are wound, as well as the wiper arm, is clearly visible.

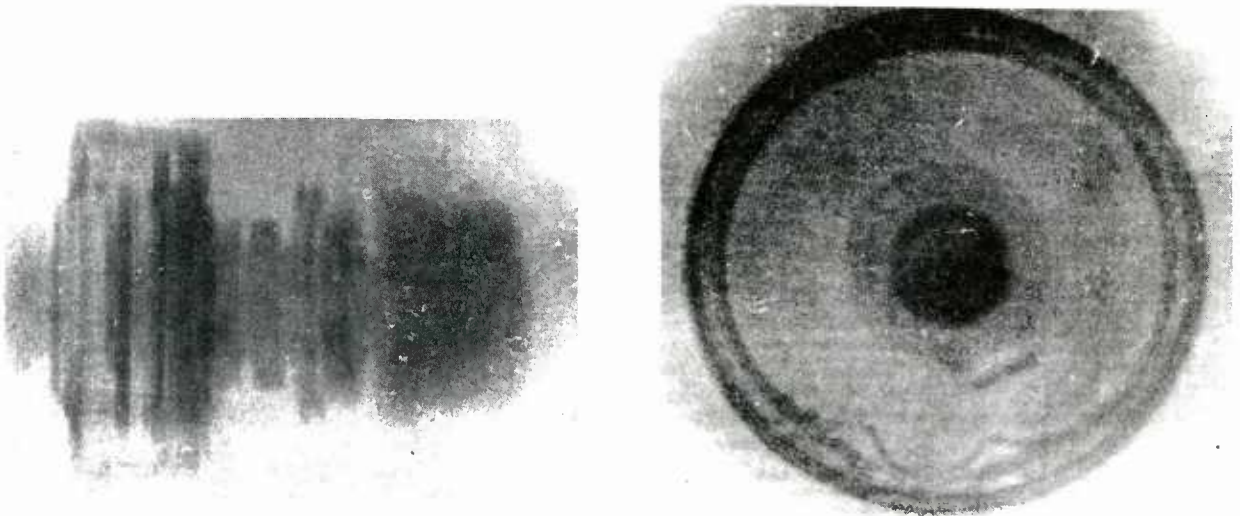


Figure 5. Fission neutron side view of a flat carbon element potentiometer (left) with plastic knob and several washers and nuts on its shaft. Case is of plastic and steel. Shaft-end view of wire-wound potentiometer in cadmium-plated steel case (right).

19-066-1213/AMC-72

Figure 6 shows two steel milling machine cutters, with one viewed from its side and the other along its axis. The axis of the cutter was not in perfect alignment with the neutron beam, causing the shaft of the cutter to cast an elongated image.

Figure 7 is a radiograph of one end of a 4-kW transmitting tube. The outer case is of ceramic, and relatively transparent to fast neutrons. The interior metal electrodes are partly conical and partly cylindrical in shape, and the outlines are very evident on the figure.

Figure 8 is an example of the ability of fission neutrons to provide images of objects through absorbers. The largest object in the composite assembly is an iron plate 13-mm thick which contains a threaded hole in its center. A smaller iron plate of the same thickness is supported on two polystyrene blocks in such a way that it covers half of the hole in the larger plate. A small rubber washer is placed so that half of it is viewed through one iron plate, and the other half is viewed through both. Note that although all components are imaged well, the radiation has penetrated both the iron and the hydrogenous materials considerably.

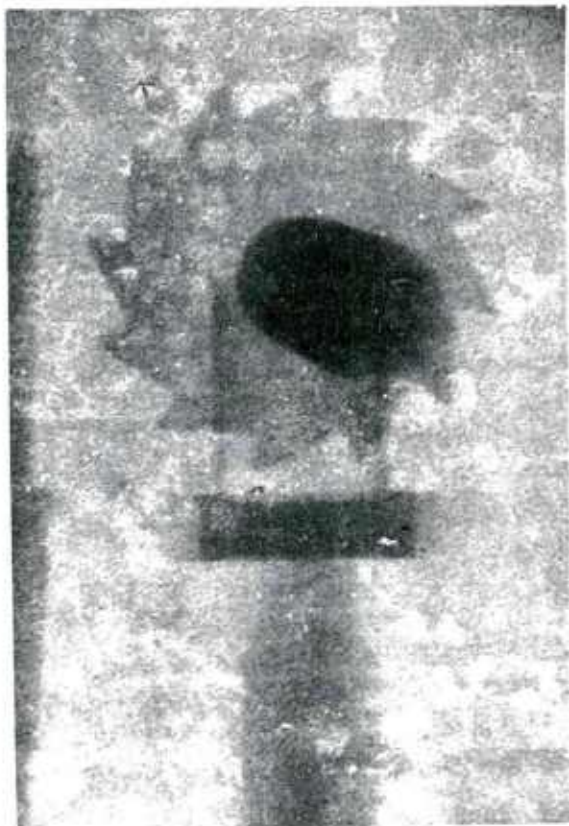


Figure 6. Fission neutron radiograph of two milling machine tools. The shaft of the upper tool is at an angle to the direction of the incident neutrons.

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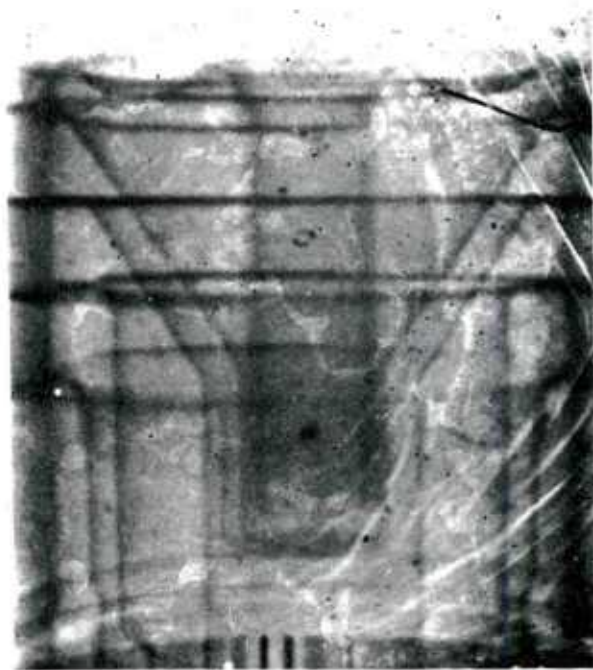


Figure 7. Fission neutron radiograph of the electrode section of a 4-kW ceramic transmitting tetrode.

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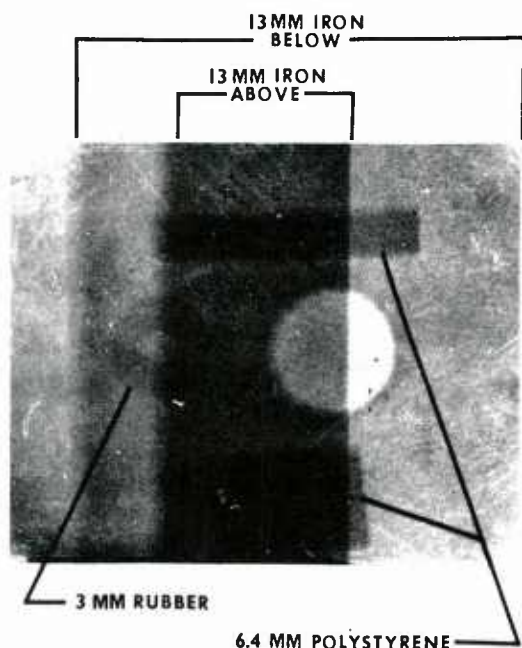


Figure 8. Demonstration of fission neutron penetration and detection capability for iron and hydrogenous materials. Lower iron plate contains a threaded hole 13-mm in diameter. The dimensions given are material thicknesses in the direction of the incident neutrons.

19-066-1207/AMC-72

CONCLUSION

The fission neutron radiography system described here is in an early stage of development, and its several attractive features encourage us to seek further improvements, particularly in increased detection efficiency. A prominent feature is its simplicity. The system requires only a bare californium point source and two inexpensive plastic films to obtain images of high resolution which are developed without darkroom procedures. The absence of collimators and moderators allows more efficient use of the available neutrons through lower absorption losses and a capability for simultaneous exposures.

Fission neutrons interact with all materials about equally well so that all combinations of materials can be examined with a good penetration capability. In this sense the system can be thought of as a general radiographic procedure similar to X-radiography which retains the ability to image materials containing even the lightest elements, as does thermal neutron radiography. The system should be immediately useful in special cases where both X-ray and thermal neutron radiography might fail, such as the radiography of highly radioactive materials and the imaging of hydrogenous components within heavily cadmium-plated containers.

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illus, D/A Project 1T161102AH42
AMCMS Code 611102.11.85500

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